Superior radiation resistance of $In_{1-x}Ga_xN$ alloys: Full-solar-spectrum photovoltaic material system

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High-efficiency multijunction or tandem solar cells based on group III–V semiconductor alloys are applied in a rapidly expanding range of space and terrestrial programs. Resistance to high-energy radiation damage is an essential feature of such cells as they power most satellites, including those used for communications, defense, and scientific research. Recently we have shown that the energy gap of $In_{1-x}Ga_xN$ alloys potentially can be continuously varied from 0.7 to 3.4 eV, providing a full-solar-spectrum material system for multijunction solar cells. We find that the optical and electronic properties of these alloys exhibit a much higher resistance to high-energy (2 MeV) proton irradiation than the standard currently used photovoltaic materials such as GaAs and GaInP, and therefore offer great potential for radiation-hard high-efficiency solar cells for space applications. The observed insensitivity of the semiconductor characteristics to the radiation damage is explained by the location of the band edges relative to the average dangling bond defect energy represented by the Fermi level stabilization energy in $In_{1-x}Ga_xN$ alloys. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1618353]

I. INTRODUCTION

Currently used multijunction solar cells are based on two or three junctions of different semiconductor materials connected in series.[1–3] Efficiencies exceeding 30% have been achieved with $Ga_{0.51}In_{0.49}P/GaAs/Ge$ triple junction devices.[4] Theoretical considerations of the solar cell efficiency have shown that incorporation of a fourth junction made of a semiconductor with an energy gap of about 1 to 1.1 eV will significantly improve the performance of the existing multijunction (MJ) cells.[5,6] One of the candidate materials for the fourth junction is the $Ga_{1-y}In_yN_xAs_{1-x}$ alloy, which with the proper choice of the composition can be grown lattice matched to GaAs and has a bandgap in the desired energy range.[7,8] There has been an intense effort aimed at fabricating four-junction solar cells by adding this material. However, so far the quality of these quaternary alloys is inferior to that of the other component materials of the existing MJ cells, and is detrimental to their overall performance.[9,10]

Recently, the energy gap of wurtzite InN has been found to be about 0.7 eV.[11–13] This gap is almost three times smaller than the previously claimed value of 2 eV.[14] This discovery has extended the range of the energy gaps of group III-nitride alloys from the deep ultraviolet to the practically very important near infrared spectral region. It has been shown that the band gap of $In_{1-x}Ga_xN$ alloys can be varied continuously from 0.7 to 3.4 eV.[15] As is shown in Fig. 1, this spectral range provides an almost perfect fit to the solar spectrum, offering a unique opportunity to design MJ solar cells using a single ternary alloy system.

Since outer space is one of the primary locations for applications of MJ solar cells, the sensitivity to high-energy particle damage is one of the most important characteristics of these materials.[16,17] It is well-known that the primary cause for the degradation of solar cells in space are due to bombardment by protons and electrons in the energy range of electron volts to hundreds of million electron volts.[14]

It has been shown that solar cell degradation in a space radiation environment can be successfully modeled using the displacement damage dose methodology.[18] The displacement damage dose (D_d) is given by the product of nonionizing energy loss (NIEL) and the particle fluence. For an incident energetic particle traversing a target, NIEL is the energy loss responsible for the production of displaced atoms and thus creating trapping and recombination centers in semiconductors. Radiation induced effects have been successfully correlated for different types of radiation (different particles and energies) on the basis of NIEL.[19] Using the D_d approach, degradation of photovoltaic parameters (such as short circuit current, open circuit voltage, and maximum power) due to proton and electron irradiation can be described by a single

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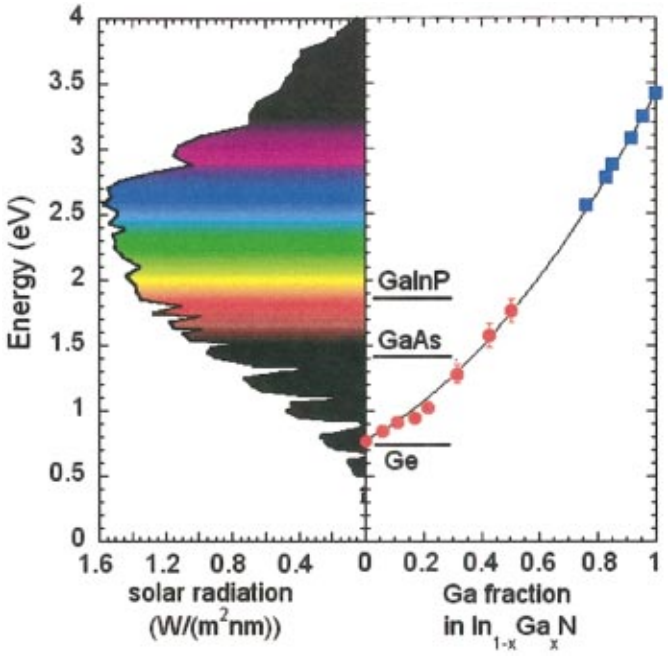

FIG. 1. (Color) Bandgap energies of the InGaN alloy system cover the entire air-mass-1.5 solar spectrum. The gap energies of conventional MJ solar cell materials (Ge, GaAs, and GaInP) are also shown in the right-hand panel for comparison.

curve for all energies and therefore can be used to accurately predict cell performance in any radiation environment based on only a limited number of ground test data.[18]

Since the NIEL can be calculated for protons with different energies in any material, this approach is particularly useful in predicting cell performance for cells fabricated using new materials. In this work, we investigate the effects of high-energy proton irradiation on the optical and electrical properties of In-rich $In_{1-x}Ga_xN$ alloys as a function of the displacement damage dose. We show side-by-side comparisons that InGaN is less sensitive to high-energy particle damage than GaAs and GaInP, the semiconductors that are currently used in MJ cells. Work on InGaN has not yet progressed to the point of making complete devices, so we have chosen to study here basic material properties. These material properties change by orders of magnitude when GaAs or GaInP is irradiated, and can be used to predict performance of solar cells and other optoelectronic devices. InGaN is shown to withstand an extraordinarily high dose of radiation. Although we expect that the InGaN would have degraded more if its initial quality were improved, after radiation it outperforms GaAs and GaInP, implying that further improvements in the initial material quality will not change the final outcome.

This exciting result motivates a substantial effort to fabricate solar cells out of the InGaN material system. Work will be needed to fabricate *p*-type InN and InGaN in the range of 1.8 to 2.5 eV, and to fabricate full devices, including tunnel junctions. In addition, we predict that InGaN-based electronics will show extraordinary radiation hardness and will be useful for a wide range of functions in high radiation applications.

II. EXPERIMENT

InN ($\sim$7.5 μm) and $In_{1-x}Ga_xN$ films ($\sim$240 nm, x up to 0.32) were grown on (0001) sapphire substrates by molecular beam epitaxy.[20] GaN and $In_{0.08}Ga_{0.92}N$ samples were grown by metalorganic chemical vapor deposition (MOCVD).[21] For comparison purposes we have also included MOCVD-grown GaAs (5 μm) and $Ga_{0.51}In_{0.49}P$ (1 μm) (Ref. 22) films in our studies. These samples were characterized by photoluminescence (PL) spectroscopy, Hall effect, and resistivity measurements at room temperature. For GaN and $In_{0.08}Ga_{0.92}N$, the PL signals were generated by excitation with the 325 nm line of a HeCd laser ($\sim$3 mW). The signals were then dispersed by a 1 m double-grating monochromator and detected by a Hamamatsu R928 photomultiplier tube. For all the other samples, the 476.5 nm line ($\sim$5 mW) of an argon laser was used as the excitation source. The photomultiplier tube, a Ge photodiode, and a liquid-nitrogen cooled InSb detector were used to detect the band-edge PL signals of $Ga_{0.51}In_{0.49}P$, GaAs, and In-rich InGaN, and InN samples, respectively. Hall effect and resistivity measurements were done using the standard van der Pauw method with a magnetic field of 3000 G.

A 2 MeV proton beam generated by a Van de Graaff accelerator was defocused to a beam size of $\sim 4\times 7$ mm^2. Since the materials we study are thin films with thickness <8 μm, the energy loss density of the 2 MeV proton beam through these films is practically constant. NIEL of 2 MeV proton in different materials was calculated by the Monte Carlo SRIM software[23] and is found to be in the range of $5.0-5.6\times 10^{-2}$ MeV/(g/cm^2) for the various target materials. Proton irradiations with doses of 0.45, 1.12, and 2.23 $\times 10^{14}$ cm^{-2} were used, corresponding to a displacement damage dose D_d in the range of 2.2–2.5, 5.5–6.2, and $11.0-12.5\times 10^{12}$ MeV/g, respectively, for the various target materials. We also note that our 2 MeV proton irradiation at a dose of 10^{14} cm^{-2} could result in a same degree of degradation in maximum power of GaAs/Ge solar cells as that of a 1 MeV electron irradiation at a dose of 5×10^{17} cm^{-2}.[18]

III. EXPERIMENTAL RESULTS AND DISCUSSION

A. Photoluminescence results

Figure 2 shows the band-edge PL spectra of an InN and an $In_{0.68}Ga_{0.32}N$ sample measured before and after the proton irradiation with different doses. For comparison, the PL spectra of GaAs and $Ga_{0.51}In_{0.49}P$ are also shown. It is obvious that the PL intensities of GaAs and $Ga_{0.51}In_{0.49}P$ are drastically suppressed by the irradiation. For example the PL peak intensity of $Ga_{0.51}In_{0.49}P$ decreases by two orders of magnitude after a proton irradiation at 1.12×10^{14} cm^{-2}. In stark contrast, the PL signal of InN does not show any reduction in intensity after being subjected to a similar radiation dose. A representative example of the effects of the irradiation on the properties of In-rich $In_{1-x}Ga_xN$ alloys is shown in Fig. 2. It is seen that the PL intensity of $In_{0.68}Ga_{0.32}N$ is reduced by about four times and the PL peak position is blue shifted by about 0.1 eV.

The PL peak intensities of these samples normalized to the corresponding pre-irradiation intensities are shown as a

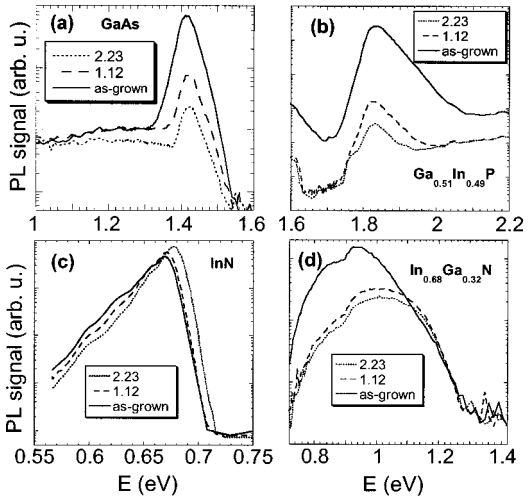

FIG. 2. Near band-edge PL spectra of GaAs, $Ga_{0.51}In_{0.49}P$, InN, and $In_{0.68}Ga_{0.32}N$ before and after proton irradiation with doses of 1.12 and 2.23×10^{14} cm^{-2}. Note that their vertical axes are in log scales covering different decade ranges.

function of D_d in Fig. 3. We note that the GaAs and $In_{0.68}Ga_{0.32}N$ samples have comparable band-edge PL intensities prior to the irradiation. It is clearly seen that, in general, nitrides are less sensitive to the irradiation as compared to GaAs and GaInP. It is widely known that GaN and Ga-rich InGaN materials maintain excellent optical properties even in the presence of high densities of structural defects. Highly efficient light emission can be easily achieved from defective GaN and Ga-rich InGaN alloys, making them the key materials used in blue light emitting diodes. Figure 3 shows that the optical properties of InN and In-rich InGaN alloys are even more remarkable. No strong reduction in PL intensity is observed in InN and $In_{1-x}Ga_xN$ with small Ga fractions. However, it is interesting to note the trend that as the Ga fraction increases, the irradiation-induced degradation in optical emission becomes increasingly severe. To compare the resistance of these materials to irradiation in a quantitative

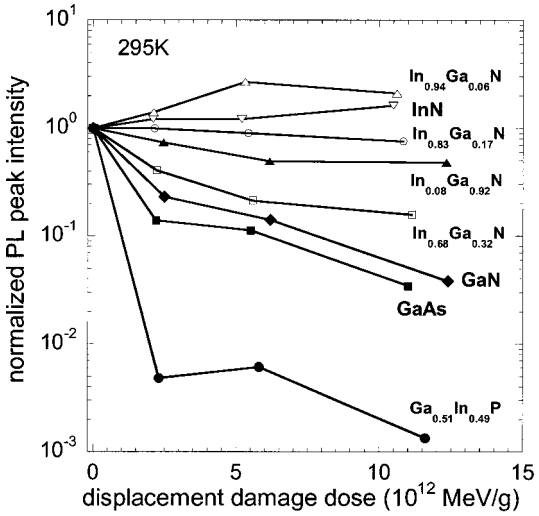

FIG. 3. Normalized PL peak intensity as a function of displacement damage dose.

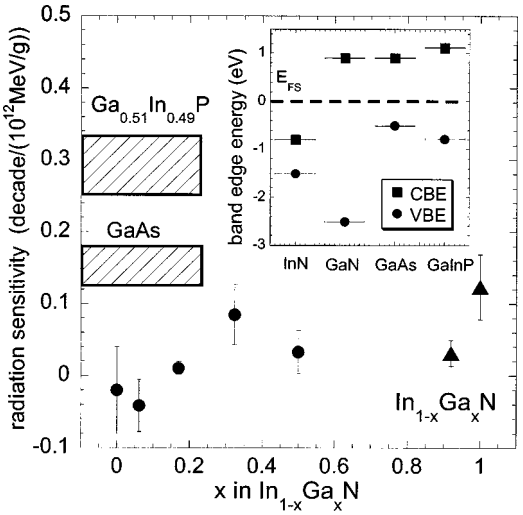

FIG. 4. Radiation sensitivity of $In_{1-x}Ga_xN$ alloys investigated. Solid circles represent In-rich InGaN alloys grown by molecular beam epitaxy under similar conditions in the same laboratory. Triangles are GaN and Ga-rich InGaN grown in another laboratory. The sensitivity level of GaAs and GaInP are also shown as shaded areas. Inset, conduction band-edge and valence band-edge alignments of InN, GaN, GaAs, and $Ga_{0.51}In_{0.49}P$ referenced to the universal Fermi stabilization level.

way, we use the magnitude of the slope of the curves shown in Fig. 3 as a measure of their radiation sensitivity. The radiation sensitivities in units of decade/(10^{12} MeV/g) for these materials are shown in Fig. 4. The error bar of each data point is given by the uncertainty of the slope when fitting the corresponding degradation curve in Fig. 3 with a straight line. It can be seen that the radiation sensitivity of GaAs and GaInP is far above that of InN, GaN, and InGaN alloys.

B. Discussion of radiation resistance

The considerably different irradiation behavior of these materials can be understood in terms of defects generation in semiconductors by high-energy particle irradiation. It is known that the energy location of a charge transition state associated with a broken bond is determined by a universal energy reference known as the Fermi stabilization level (E_{FS}).[24,25] This level defines the average dangling bond energy with respect to band edges in semiconductors. For example, it was found that E_{FS} lies $\sim$0.9 eV below the conduction band edge in GaN.[26] Assuming that the defects generated by high-energy proton irradiation are of broken bond character, we can use E_{FS} to quantitatively predict the energy locations of the defect levels in various semiconductors. Using the known values of band offsets between InN, GaN, GaAs, and GaInP,[27–29] we illustrate in the inset of Fig. 4 the location of E_{FS} with respect to the band edges of these materials. It can be seen that E_{FS} lies inside the conduction band of InN, while for GaN, GaAs, and GaInP it falls into the band gap. Indeed, it has been reported by using deep level transient spectroscopy and positron annihilation spectroscopy that high-energy proton irradiation on GaInP creates deep defects with energy level located near mid band gap.[30]

It has been recognized that defect levels located close to the midgap position are the most effective nonradiative recombination centers.[24,25,31] This explains the strong effect of the irradiation on the PL of GaInP where E_{FS} is located close to the mid-gap. It has been also noted that nonradiative recombination rates decrease in semiconductors such as InP, in which E_{FS} lies closer to the conduction band edge.[24,25]

An extreme situation is represented by semiconductors in which the defect levels lie inside the conduction or the valence bands. Such is the case for InN, in which E_{FS} is located at ~ 0.8 eV (more than the gap energy of InN) above the conduction band edge. High-energy barriers for the carrier recombination through the dangling bond-like defect states prevents those defects from acting as efficient recombination centers, resulting in a spectacular insensitivity of InN to high-energy particle irradiation. On the other hand, irradiation-generated defects provide fast nonradiative recombination centers in GaAs and GaInP and effectively quench the photoluminescence.

The situation for GaN lies between these two extreme cases. The behavior of InGaN alloys is intermediate between that of InN and GaN. In Fig. 4 it can be seen that the In-rich InGaN alloys, which were grown in the same laboratory under similar conditions, show an increasing sensitivity to the irradiation with increasing Ga fraction. This trend reflects the upward motion of E_{FS} toward the conduction band edge of InGaN. Using linear interpolation, it can be estimated that the conduction band edge of $In_{1-x}Ga_xN$ crosses E_{FS} at $x \sim 0.47$. For the Ga-rich $In_{0.08}Ga_{0.92}N$ film that was grown in a different laboratory, the data do not follow the trend of the In-rich InGaN set, but are still within the range between InN and GaN.

Although PL intensity is only one of the semiconductor characteristics that relates to the solar cell performance, it has been shown that in the case of GaAs/Ge cells the maximum power is reduced by about 90% after a proton irradiation with $D_d = 2 \times 10^{12}$ MeV/g.[32] Such a performance degradation agrees well with the reduction in the PL intensity of GaAs for comparable D_d as shown in Fig. 3. Therefore, it is reasonable to assume that the insensitivity of the optical properties of InGaN alloys to the radiation damage is a good indicator of expected radiation hardness of photovoltaic devices made with these *n*-type alloys. The radiation resistance of complementary *p*-type materials must also be explored before the radiation resistance of complete InGaN photovoltaic devices can be predicted.

C. Photoluminescence lifetime

To further assess the effect of the irradiation on the generation of nonradiative recombination centers, we have measured the PL lifetime using time-resolved photoluminescence technique. PL decay curves were measured by time-correlated single-photon counting[33] at room temperature. The excitation source was a 1-MHz pulse train at 580 nm with an average power of 5 mW and a spot size of about 1 mm. The time resolution of the experiment was about 30 ps for the GaInP samples, and 100 ps for the InGaN samples. The average PL decay time was determined by the formula

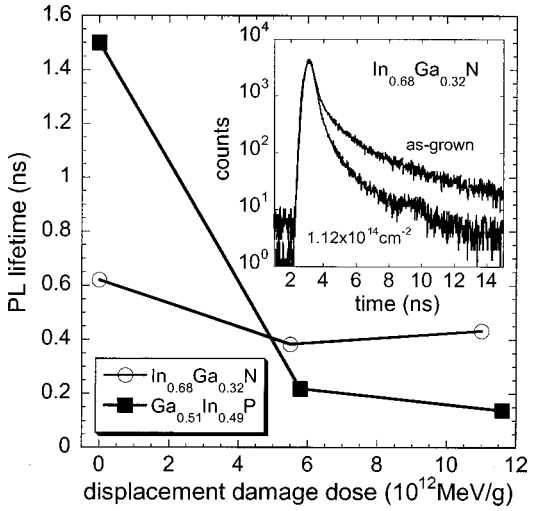

FIG. 5. Photoluminescence lifetimes of $Ga_{0.51}In_{0.49}P$ and $In_{0.68}Ga_{0.32}N$ as functions of displacement damage dose. Inset shows the integrated PL decay curves of $In_{0.68}Ga_{0.32}N$ before and after 1.12×10^{14} cm^{-2} proton irradiation. These curves are rescaled to equal peak intensity.

$$\tau = \sum_{m=\text{initial}}^{\text{final}} (N_{m-1} - N_m)(t_m - t_{\text{initial}})/(N_{\text{initial}} - N_{\text{final}}), \tag{1}$$

where N_m represents the number of photon counts in channel m, and t_m represents the time corresponding to that channel. The function is applied over several orders of magnitude of decay, and gives a lifetime similar to what one attains by averaging the lifetimes, weighted by the prefactors, of multiexponential fits. Generally, the initial decay dominates the value for the lifetime..

Figure 5 shows the lifetime of the PL signals of $Ga_{0.51}In_{0.49}P$ and $In_{0.68}Ga_{0.32}N$. In the inset of Fig. 5, representative PL decay curves of $In_{0.68}Ga_{0.32}N$ are shown. For un-irradiated samples the PL lifetime of $Ga_{0.51}In_{0.49}P$ is 2.5 times longer the lifetime of $In_{0.68}Ga_{0.32}N$. However it is seen in Fig. 5 that the radiation damage has a dramatic effect on the lifetime of $Ga_{0.51}In_{0.49}P$ with the lifetime reduced by an order of magnitude for the highest irradiation dose. In stark contrast, $In_{0.68}Ga_{0.32}N$ is found to be much less sensitive to the radiation damage. The highest radiation dose reduces the PL lifetime by only 50% to about 0.4 ns, which is more than two times longer than the PL lifetime of $Ga_{0.51}In_{0.49}P$ after similar irradiation. This result is consistent with the PL measurements that show a much smaller effect of radiation damage on the PL intensity of $In_{0.68}Ga_{0.32}N$ than on that of $Ga_{0.51}In_{0.49}P$.

D. Transport properties

The observed correlation between the energy position of E_{FS} relative to the band edges and the effects of radiation on the material properties indicates that the radiation hardness of InN and InGaN alloys is an intrinsic property of these materials, rather than an extrinsic insensitivity caused by the high density of defects that already exist in the starting materials. This explanation is further supported by the effects of irradiation on the transport properties of these materials.

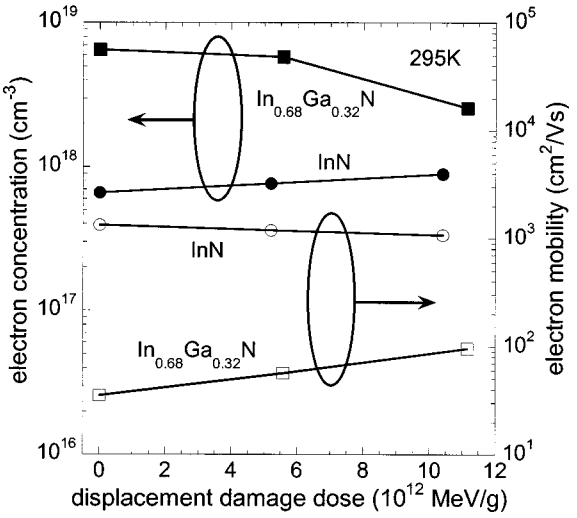

FIG. 6. Free electron concentration and mobility of InN and $In_{0.68}Ga_{0.32}N$ as a function of displacement damage dose measured by the Hall effect.

Prior to the irradiation, the GaAs film is *n*-type with a resistivity of 146 Ω cm. After the irradiation with 1.12×10^{14} cm^{-2} of protons, it becomes highly resistive with resistivity exceeding 10^4 Ω cm. A similar increase in resistivity after irradiation has been reported for GaInP (Ref. 20) and also observed in our GaInP films. On the other hand, as demonstrated in Fig. 6, InN shows only a small increase (about 15%) in the free electron concentration and an 11% reduction in the free electron mobility, corresponding to a negligible change in resistivity from 0.0071 to 0.0069 Ω cm. $In_{0.68}Ga_{0.32}N$ shows a slight decrease in free electron concentration and a slight increase in mobility, corresponding to a decrease in resistivity from 0.027 to 0.019 Ω cm. This striking difference indicates that the irradiation-generated defect states are efficient electron traps in GaAs and GaInP, while in InN and In-rich InGaN they are not. The robustness of the optical and transport properties against irradiation for the InN-based materials is highly desirable for their potential applications in photovoltaic and other devices operated in hostile space environments. We emphasize that the radiation levels that were tolerated by the InGaN materials are an order of magnitude higher than those that are typically applied to space cells. Thus, these results imply that InGaN solar cells could be used in high-radiation orbits that have previously been unapproachable.

IV. CONCLUSIONS

In conclusion, photoluminescence spectroscopy, Hall effect, and resistivity measurements have been performed on InN and InGaN films after high-energy proton irradiation. Compared with conventional solar cell materials (GaAs and GaInP), InN and $In_{1-x}Ga_xN$ alloys show much less deterioration in their optical and transport properties. The different behaviors are explained in terms of the different energy configurations of the irradiation-induced defect states in the band diagram of the host materials. Given the fact that the band-gap energy of the $In_{1-x}Ga_xN$ alloys is tunable between 0.7 and 3.4 eV covering the full solar spectrum and their superior resistance against irradiation damage $In_{1-x}Ga_xN$ alloys present a great potential for applications in photovoltaic and other optoelectronic devices, motivating further development of these materials.

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